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Coulomb Interactions in an Atomic Dielectric*

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The Coulomb interaction between two impurity charges is evaluated for an atomic dielectric. The point-dipole model is used to describe the model dielectric. The interaction potential between the two impurity charges may be expressed as an asympotic series in $1/R$, where R is the separation of the two charges. We derive a method for systematically evaluating the coefficients of this series, and explicitly calculate all terms up to and including those for $1/R^3$ for the monatomic cubic lattices. The leading term is $1/\epsilon R$.

I. INTRODUCTION

In 1837 Michael Faraday discovered that the difference between vacuum and an insulator can, for the purposes of electrostatics, be characterized by a single number, called by him the *specific inductive capacity* of the insulator. Nowadays, it is more commonly called the dielectric constant.[1] As every schoolchild now knows, the force between two charges in a medium is reduced from that in vacuum by a factor $1/\epsilon$, where ϵ is the dielectric constant.

A little reflection on modern views of the constitution of matter led us to suspect that this is an asymptotic law, valid for large distances between the charges compared to distances characterizing the microscopic structure of the insulator. We therefore have investigated the leading corrections to Coulomb's law (modified by $1/\epsilon$) in crystalline solids. Naturally, we did not expect, nor did we find, effects which would cause any modification of conventional macroscopic electrostatics. However, even in microscopic calculations in solid-state physics, the macroscopic idea of a dielectric constant is used, and corrections such as we discuss may be relevant. Such problems might be, for example, Wannier excitons in solids,[2] or the off-center location of Li^+ impurities in alkali-halide crystals.[3] In this paper, we do not consider specific applications, but address ourselves to the basic question of the interaction of impurity charges in an insulating crystal.

What we do is clearly equivalent, in some sense, to computing the longitudinal nonlocal dielectric function $\epsilon(k)$, though we have not formulated the problem in this way at all. Our method, we believe, keeps the physical origins of the various terms well to the fore. The physical picture is that the impurity charges induce polarization clouds in the crystal, and these induced moments interact in a manner also conditioned by the presence of the crystal. One can see, in our formalism, which terms are due to the induction and which to the interaction.

The calculation proceeds from a purely microscopic point of view. We put two impurity charges into the dielectric, and calculate the interaction between them. Each impurity atom may be anywhere within its unit cell of the crystal. We assume that the positions of all host atoms are unchanged by the presence of the impurities; thus our impurities are interstitial rather than substitutional. In order to make the calculation tractable, we have adopted the point-dipole model for the host dielectric. This model assumes that the induced polarization moments of the host ions may be represented by point dipoles located at the atomic site. The two impurity charges polarize these host atoms, thereby affecting the net interaction between them. One of us has previously derived the formula describing this interaction.[4] Now we wish to show how to evaluate this interaction. Specifically, this interaction can be expanded in an asymptotic series in powers of $1/R$, where R is the separation of the two impurities. We derive a method for systematically evaluating the coefficients of this series, and explicitly calculate all terms up to and including those for $1/R^3$ for simple cubic sc, fcc, and bcc lattices.

This problem has received only scant attention in the literature. Most discussions of the corrections to the $1/\epsilon R$ potential have been concerned with dynamic effects caused by the motion of the charges: An example is Haken's calculation for excitons.[5] In contrast, our impurity charges are fixed, and the deviations from $1/\epsilon R$ arise merely from the atomicity of the dielectric. The only work which appears directly related to ours is Sham's derivation of the potential an electron feels near a donor atom in a semiconductor.[6] Sham's approach differs from ours in many respects. He gets dynamic effects from letting the electron move, he only calculates $1/R^2$ terms, while we go to higher order, he includes correlation and exchange effects, and he derives his results by many-body theory. Although his approach is more general than ours, it has the drawback that the

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[1] Henry Cavendish had anticipated this idea, but this remained unknown until his papers were edited by J. C. Maxwell in 1879. See E. T. Whittaker, *A History of the Theories of Aether and Electricity* (Thos. Nelson and Sons, London, 1951), Vol. 1.

[2] For a recent review, see B. Segall and D. T. F. Marple, in *Physics and Chemistry of II-VI Compounds*, edited by M. Aven and J. S. Prener (North-Holland Publishing Co., Amsterdam, 1967).

[3] See, for example, W. D. Wilson, R. D. Hatcher, G. J. Dienes, and R. Smoluchowski, Phys. Rev. **161**, 888 (1967).

[4] G. D. Mahan, Phys. Rev. **153**, 983 (1967); this article is referred to as I.

[5] H. Haken, in *Polarons and Excitons*, edited by C. G. Kuper and G. D. Whitfield (Plenum Press, Inc., New York, 1963).

[6] L. J. Sham, Phys. Rev. **150**, 720 (1966).

constant parameters which arise are not readily calculated from a microscopic model. The advantage of the point-dipole model is that all of the expansion constants can be quickly calculated from the lattice constants and atomic polarizabilities. Furthermore, the point-dipole system is a clean mathematical model well worth investigating.

In Sec. II we describe and justify the method of generating an expansion in $1/R$. The leading coefficients are evaluated in Sec. III, and the results are discussed in Sec. IV. Many of the mathematical details are relegated to the several Appendices.

II. SURFACE INTEGRALS AND SUBTRACTIONS

A. Statement of the Problem

In a previous paper[4] on the subject of local field corrections (denoted by I), one of us has given a formula for the potential energy of interaction of two impurities with charges q_1 and q_2 at positions $\mathbf{l}_1$ and $\mathbf{l}_2$ in a crystal, the separation between them being denoted by $\mathbf{R}=\mathbf{l}_1-\mathbf{l}_2$. Each of the positions $\mathbf{l}_1$ and $\mathbf{l}_2$ could be anywhere in the unit cell, but we do assume that all of the host ions are present. In this case it was shown in I that the effective interaction between these two point impurities is

$$V(\mathbf{l}_1,\mathbf{l}_2)=q_1q_2\left[\frac{1}{R}-4\pi\alpha\int_{\mathrm{BZ}}\frac{d^3k}{2\pi^2}e^{i\mathbf{k}\cdot\mathbf{R}}f(\mathbf{k};\mathbf{l}_1,\mathbf{l}_2)\right]. \quad (2.1)$$

The notation BZ means the integration volume is confined to the Brillouin zone. Similarly, we shall use OBZ for integrations which include all wave vector space outside of the Brillouin zone. The function f is given by

$$f(\mathbf{k};\mathbf{l}_1,\mathbf{l}_2)=W_\mu(\mathbf{l}_1,\mathbf{k})G_{\mu\nu}(\mathbf{k})W_\nu(\mathbf{l}_2,-\mathbf{k}), \quad (2.2)$$

where the vector function W_μ is given by

$$W_\mu(\mathbf{l}_1,\mathbf{k})=\frac{V_0}{4\pi}\sum_{\mathbf{l}}e^{i\mathbf{k}\cdot(\mathbf{l}-\mathbf{l}_1)}\frac{(\mathbf{l}-\mathbf{l}_1)_\mu}{(\mathbf{l}-\mathbf{l}_1)^3}. \quad (2.3)$$

We use the summation convention for indices. The sum in (2.3) extends over all lattice sites $\mathbf{l}$, and V_0 is the volume of a unit cell. The tensor $G_{\mu\nu}(\mathbf{k})$ is obtained by first evaluating a tensor $T_{\mu\nu}(\mathbf{k})$ which is also defined as a lattice sum:

$$T_{\mu\nu}(\mathbf{k})=\frac{V_0}{4\pi}\sum_{l\neq0}e^{i\mathbf{k}\cdot\mathbf{l}}\phi_{\mu\nu}(l),$$
$$\phi_{\mu\nu}(l)=(1/l^3)[\delta_{\mu\nu}-3l_\mu l_\nu l^{-2}], \quad (2.4)$$

and then solving

$$[\delta_{\mu\nu}+4\pi\alpha T_{\mu\nu}(\mathbf{k})]G_{\nu\lambda}(\mathbf{k})=\delta_{\mu\lambda}. \quad (2.5)$$

In I it is stated that if $\hat{\epsilon}_1$, $\hat{\epsilon}_2$, and $\hat{\epsilon}_3=\hat{k}$ are a set of appropriately chosen orthonormal unit vectors, then it follows that in cubic crystals

$$\hat{\epsilon}_i\cdot\mathbf{T}\cdot\hat{\epsilon}_j=\delta_{ij}T_i(k), \quad (2.6)$$

and this led to a simple expression for $G_{\mu\nu}(\mathbf{k})$. Unfortunately, (2.6) is incorrect for an arbitrary wave vector $\mathbf{k}$. This means that after evaluating $T_{\mu\nu}(\mathbf{k})$ one must in general invert the matrix equation (2.5) to find $G_{\mu\nu}(\mathbf{k})$.[7] The simple result (2.6) was mistakenly thought to be correct because it is valid in cubic crystals at $k\to0$ and also along the symmetry directions (100), (110), and (111). Since (2.6) is valid as $k\to0$, then the asymptotic results ($R\to\infty$) derived in Ref. 1 are still correct.

In I, Eq. (2.1) was evaluated by making the approximations of considering only the lowest-order contribution to f, and carrying the integration over all space, instead of the Brillouin zone. In the following we show how these approximations may be improved.

Equations (2.1) to (2.5) just apply to lattices with one atom per unit cell. When there are more than one atom per unit cell, as in the diamond or NaCl structure, one must solve a set of coupled equations in order to obtain the effective interaction. As an example, the solution for two atoms per unit cell is given in Appendix A. For simplicity, we are going to restrict our present discussion to just cubic lattice with one atom per unit cell—i.e., sc, bcc, and fcc structures. This has already been anticipated since we have assumed that the atomic polarizability α is isotropic in deriving (2.1) and (2.5).

We should emphasize that $f(\mathbf{k};\mathbf{l}_1,\mathbf{l}_2)$ does not depend significantly upon the separation R. This is because $W_\mu(\mathbf{l}_1,\mathbf{k})$ is a periodic function of $\mathbf{l}_1$, and hence has the same value in each atomic cell. The value of W_μ does depend upon the position of the impurity q_1 at $\mathbf{l}_1$ within the unit cell. Thus the function $f(\mathbf{k};\mathbf{l}_1,\mathbf{l}_2)$ depends upon where each impurity at $\mathbf{l}_1$ and $\mathbf{l}_2$ are in their respective unit cells, but it does not depend upon the separation of these two unit cells. The separation of unit cells is entirely expressed by the R factor in the exponent of (2.1).

B. Surface Integrals

Our aim is to evaluate (2.1) as a power series in R^{-1}. This will probably be an asymptotic series, and the first few terms will provide an accurate representation of $V(\mathbf{l}_1,\mathbf{l}_2)$ as R becomes large. We generate this series by changing the volume integral in (2.1) to a surface integral over the Brillouin zone.

The trick we use to generate surface integrals starts with the identity

$$e^{i\mathbf{k}\cdot\mathbf{R}}=\frac{\mathbf{R}\cdot\nabla_k}{iR^2}(e^{i\mathbf{k}\cdot\mathbf{R}}). \quad (2.6')$$

When we put (2.6′) into the integrand of (2.1), and

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[7] That is, Eq. (13) of Ref. 4 is incorrect for an arbitrary wave vector.

integrate once by parts, we get

$$\int_{\mathrm{BZ}} \frac{d^3k}{2\pi^2} e^{i\mathbf{k}\cdot\mathbf{R}} f = \frac{1}{2\pi^2 i R^2} \oint d\mathbf{S}\cdot\mathbf{R} e^{i\mathbf{k}\cdot\mathbf{R}} f + \frac{i}{2\pi^2 R^2} \int_{\mathrm{BZ}} d^3k\, e^{i\mathbf{k}\cdot\mathbf{R}} \mathbf{R}\cdot\nabla_k f. \quad (2.7)$$

The surface integral runs over the Brillouin-zone surface. Now, we can repeat this trick on the remaining volume integral in (2.7). In fact, after repeating the trick n times we get

$$\frac{1}{2\pi^2} \int_{\mathrm{BZ}} d^3k\, e^{i\mathbf{k}\cdot\mathbf{R}} f = \frac{1}{2\pi^2 i R^2} \oint \mathbf{dS}\cdot\mathbf{R} e^{i\mathbf{k}\cdot\mathbf{R}} \sum_{j=0}^{n-1} \left(\frac{i\mathbf{R}\cdot\nabla_k}{R^2}\right)^j f + \frac{1}{2\pi^2} \int_{\mathrm{BZ}} d^3k\, e^{i\mathbf{k}\cdot\mathbf{R}} \left(\frac{i\mathbf{R}\cdot\nabla_k}{R^2}\right)^n f. \quad (2.8)$$

Equation (2.8) is a method of generating a power series in R^{-1} as n assumes successively higher values. The volume integral acts as a remainder term for the series, and this term is of order R^n.

We must make two important modifications to Eq. (2.8) before it can be used to generate the desired expansion in powers of R^{-1}. The first arises from the fact that $f(\mathbf{k}; \mathbf{l}_1,\mathbf{l}_2)$ is not an analytic function of $\mathbf{k}$, and this causes the volume integrals in (2.8) to diverge at $k \to 0$. This difficulty can be eliminated by subtracting from f its nonanalytic parts. The second modification of (2.8) is a simplification: All of the surface integrals vanish.

We will now show that all of the surface integrals vanish. They vanish because the surface integration over two opposing faces of the Brillouin zone exactly cancel. The normal vector to a Brillouin-zone face is given by $\mathbf{k}_0 = \frac{1}{2}\mathbf{K}_0$, where $\mathbf{K}_0$ is the appropriate reciprocal-lattice vector. For the $j=0$ surface integral term in (2.8), the integration over two opposing faces is

$$\frac{\mathbf{k}_0\cdot\mathbf{R}}{2\pi^2 i R^2} \int d^2k_\perp e^{i\mathbf{k}_\perp\cdot\mathbf{R}} [e^{i\mathbf{k}_0\cdot\mathbf{R}} f(\mathbf{k}_0,\mathbf{k}_\perp; \mathbf{l}_1,\mathbf{l}_2) - e^{-i\mathbf{k}_0\cdot\mathbf{R}} f(-\mathbf{k}_0,\mathbf{k}_\perp; \mathbf{l}_1,\mathbf{l}_2)]. \quad (2.9)$$

The $\mathbf{k}_\perp$ wave vector runs over the zone face perpendicular to $\mathbf{k}_0$. In order to show that (2.9) vanishes, we need to evaluate $f(\mathbf{k}+\mathbf{K}; \mathbf{l}_1,\mathbf{l}_2)$. First, note that if $\mathbf{K}$ is any reciprocal-lattice vector, then from (2.3)

$$W_\mu(\mathbf{l}_1, \mathbf{k}+\mathbf{K}) = \frac{V_0}{4\pi} \sum_{l} e^{i(\mathbf{k}+\mathbf{K})\cdot(\mathbf{l}-\mathbf{l}_1)} \frac{(\mathbf{l}-\mathbf{l}_1)_\mu}{(\mathbf{l}-\mathbf{l}_1)^3} = e^{-i\mathbf{K}\cdot\mathbf{l}_1} W_\mu(\mathbf{l}_1,\mathbf{k}).$$

Both $T_{\mu\nu}(\mathbf{k})$ and $G_{\mu\nu}(\mathbf{k})$ are periodic functions of reciprocal-lattice vectors. Therefore, from the definition of f in (2.2) it follows that

$$f(\mathbf{k}+\mathbf{K}; \mathbf{l}_1,\mathbf{l}_2) = e^{-i\mathbf{K}\cdot\mathbf{R}} f(\mathbf{k}; \mathbf{l}_1,\mathbf{l}_2). \quad (2.10)$$

Recalling that $2\mathbf{k}_0 = \mathbf{K}_0$, the two terms in (2.9) cancel since

$$e^{-i\mathbf{k}_0\cdot\mathbf{R}} f(-\mathbf{k}_0,\mathbf{k}_\perp; \mathbf{l}_1,\mathbf{l}_2) = e^{-i\mathbf{k}_0\cdot\mathbf{R}} [e^{2i\mathbf{k}_0\cdot\mathbf{R}} f(\mathbf{k}_0,\mathbf{k}_\perp; \mathbf{l}_1,\mathbf{l}_2)] = e^{i\mathbf{k}_0\cdot\mathbf{R}} f(\mathbf{k}_0,\mathbf{k}_\perp; \mathbf{l}_1,\mathbf{l}_2).$$

Thus we have shown that the surface integrals vanish in (2.8) for the $j=0$ term. The terms for $j \neq 0$ also vanish, and the proof is the same as for the $j=0$ terms. From (2.10) we find that

$$\left(\frac{i\mathbf{R}\cdot\nabla_k}{R^2}\right)^j f(\mathbf{k}+\mathbf{K}; \mathbf{l}_1,\mathbf{l}_2) = e^{-i\mathbf{K}\cdot\mathbf{R}} \left(\frac{i\mathbf{R}\cdot\nabla_k}{R^2}\right)^j f(\mathbf{k}; \mathbf{l}_1,\mathbf{l}_2),$$

so that all of the surface integrals over two opposing faces cancel. Since the Brillouin zone is composed of pairs of opposing faces, then

$$\oint d\mathbf{S}\cdot\mathbf{R} e^{i\mathbf{k}\cdot\mathbf{R}} \left(\frac{i\mathbf{R}\cdot\nabla_k}{R^2}\right)^j f(\mathbf{k}; \mathbf{l}_1,\mathbf{l}_2) = 0. \quad (2.11)$$

This result is valid for all values of j. The results (2.8) and (2.11) are the main results of this section.

C. Subtractions

It was noted above that the function $f(\mathbf{k}; \mathbf{l}_1,\mathbf{l}_2)$ is not an analytic function of $\mathbf{k}$. At the point $\mathbf{k}=0$ in the Brillouin zone the function f cannot be expanded in a power series in terms of the variables (k_x,k_y,k_z). For example, the volume integral on the right-hand side of (2.7) and (2.8) actually diverges, since $f \to Gk^{-2}$ as $k \to 0$, where $G \equiv (\epsilon+2)/3\epsilon$. We might be able to eliminate this divergence by subtracting the Gk^{-2} term from f:

$$\frac{1}{2\pi^2} \int_{\mathrm{BZ}} d^3k\, e^{i\mathbf{k}\cdot\mathbf{R}} f = \frac{1}{2\pi^2} \int_{\mathrm{BZ}} d^3k e^{i\mathbf{k}\cdot\mathbf{R}} (f - Gk^{-2}) + G \int_{\mathrm{BZ}} d^3k \frac{1}{2\pi^2 k^2} e^{i\mathbf{k}\cdot\mathbf{R}}.$$

If $f - Gk^{-2}$ were now an analytical function of k, we could change it into a surface integral by using (2.8). Actually, $f - Gk^{-2}$ is still not an analytic function of k, and after several operations by $(\mathbf{R}\cdot\nabla_k)$ in (2.8) we again find terms $\sim k^{-2}$ occurring. Thus we need to subtract from f an additional function besides Gk^{-2}. Let us call the total subtracted function $d(\mathbf{k}; \mathbf{l}_1,\mathbf{l}_2)$:

$$\frac{1}{2\pi^2} \int_{\mathrm{BZ}} d^3k\, e^{i\mathbf{k}\cdot\mathbf{R}} f = \frac{1}{2\pi^2} \int_{\mathrm{BZ}} d^3k\, e^{i\mathbf{k}\cdot\mathbf{R}} [f - d] + \frac{1}{2\pi^2} \int_{\mathrm{BZ}} d^3k\, e^{i\mathbf{k}\cdot\mathbf{R}} d. \quad (2.12)$$

The function $d(\mathbf{k}; \mathbf{l}_1,\mathbf{l}_2)$ will also depend upon where the impurities $\mathbf{l}_1$ and $\mathbf{l}_2$ are in their respective unit cells, but not upon the separation of unit cells. It is convenient to manipulate the last term in (2.12) by letting the integration over k go to infinity and then subtracting out the part for OBZ:

$$\int_{\mathrm{BZ}} \frac{d^3k}{2\pi^2} e^{i\mathbf{k}\cdot\mathbf{R}} d = \int_{\infty} \frac{d^3k}{2\pi^2} e^{i\mathbf{k}\cdot\mathbf{R}} d - \int_{\mathrm{OBZ}} \frac{d^3k}{2\pi^2} e^{i\mathbf{k}\cdot\mathbf{R}} d .$$

The integral to infinity is just the Fourier transform of $d(\mathbf{k};\mathbf{l}_1,\mathbf{l}_2)$, which we call $D(\mathbf{R};\mathbf{l}_1,\mathbf{l}_2)$;

$$D(\mathbf{R};\mathbf{l}_1,\mathbf{l}_2) = \frac{1}{2\pi^2} \int_{\infty} d^3k\, e^{i\mathbf{k}\cdot\mathbf{R}} d(\mathbf{k};\mathbf{l}_1,\mathbf{l}_2) . \tag{2.13}$$

The notation $D(\mathbf{R};\mathbf{l}_1,\mathbf{l}_2)$ means that D depends upon the separation R, and also upon the location of the two impurities in their unit cells. The terms in (2.12) may be collected as

$$\frac{1}{2\pi^2} \int_{\mathrm{BZ}} d^3k\, e^{i\mathbf{k}\cdot\mathbf{R}} f = D(\mathbf{R};\mathbf{l}_1,\mathbf{l}_2) + \frac{1}{2\pi^2} \int_{\mathrm{BZ}} d^3k\, e^{i\mathbf{k}\cdot\mathbf{R}} [f-d] - \frac{1}{2\pi^2} \int_{\mathrm{OBZ}} d^3k\, e^{i\mathbf{k}\cdot\mathbf{R}} d . \tag{2.14}$$

In deriving (2.14) we have made the significant assumption that the integral in (2.13) converges. We also make the assumption that d is analytic in $\mathbf{k}$ in the region OBZ. This allows us to change the last term in (2.14) into a sum of surface integrals over the Brillouin zone by using (2.8). Of course, since $f-d$ is now an analytic function of k for k within the BZ, this term can also be changed into a sum of n surface integrals by using (2.8).

$$\begin{aligned} \frac{1}{2\pi^2} \int_{\mathrm{BZ}} d^3k\, e^{i\mathbf{k}\cdot\mathbf{R}} f = {} & D(\mathbf{R};\mathbf{l}_1,\mathbf{l}_2) \\ & + \frac{1}{2\pi^2 i R^2} \oint d\mathbf{S}\cdot\mathbf{R} e^{i\mathbf{k}\cdot\mathbf{R}} \sum_{j=0}^{n-1} \left[\frac{i\mathbf{R}\cdot\nabla_k}{R^2}\right]^j (f-d) \\ & - \frac{1}{2\pi^2 i R^2} \oint d\mathbf{S}'\cdot\mathbf{R} e^{i\mathbf{k}\cdot\mathbf{R}} \sum_{j=0}^{n-1} \left(\frac{i\mathbf{R}\cdot\nabla_k}{R^2}\right)^j d \\ & + \frac{1}{2\pi^2} \int_{\mathrm{BZ}} d^3k e^{i\mathbf{k}\cdot\mathbf{R}} \left(\frac{i\mathbf{R}\cdot\nabla_k}{R^2}\right)^n (f-d) \\ & - \frac{1}{2\pi^2} \int_{\mathrm{OBZ}} d^3k e^{i\mathbf{k}\cdot\mathbf{R}} \left(\frac{i\mathbf{R}\cdot\nabla_k}{R^2}\right)^n d . \end{aligned} \tag{2.15}$$

The vector $d\mathbf{S}$ points outward from the Brillouin zone and is normal to the faces. Similarly, the vector $d\mathbf{S}' = -d\mathbf{S}$ points inward since it was derived from a volume integral for OBZ. Thus the two surface integrals of d cancel. We have already shown in (2.11) that the surface integrals of f vanish, and (2.15) immediately simplifies to

$$\begin{aligned} \frac{1}{2\pi^2} \int_{\mathrm{BZ}} d^3k\, f e^{i\mathbf{k}\cdot\mathbf{R}} = {} & D(\mathbf{R};\mathbf{l}_1,\mathbf{l}_2) \\ & + \frac{1}{2\pi^2} \int_{\mathrm{BZ}} d^3k\, e^{i\mathbf{k}\cdot\mathbf{R}} \left(\frac{i\mathbf{R}\cdot\nabla_k}{R^2}\right)^n (f-d) \\ & - \frac{1}{2\pi^2} \int_{\mathrm{OBZ}} d^3k\, e^{i\mathbf{k}\cdot\mathbf{R}} \left(\frac{i\mathbf{R}\cdot\nabla_k}{R^2}\right)^n d . \end{aligned} \tag{2.16}$$

The result (2.16) shows that we have achieved a considerable simplification in formulating our calculation. The problem of evaluating $V(\mathbf{l}_1,\mathbf{l}_2)$ in (2.1) as $R \rightarrow \infty$ has been now made relatively easy. Instead of integrating the function $f \exp(i\mathbf{k}\cdot\mathbf{R})$ over a finite volume of wave-vector space (BZ), all we need is the integration of $d(\mathbf{k};\mathbf{l}_1,\mathbf{l}_2) \exp(i\mathbf{k}\cdot\mathbf{R})$ over all k space in (2.13). This latter integration is in practice much easier to perform. As we let $n \rightarrow \infty$, the two volume integrals for BZ and OBZ become unimportant. Of course, the result (2.16) is only sensible if $D(\mathbf{R};\mathbf{l}_1,\mathbf{l}_2)$ is a uniquely defined function for large R.

One does not need to be clairvoyant to guess the total form of $d(\mathbf{k};\mathbf{l}_1,\mathbf{l}_2)$. Instead, it is possible to generate d in a step-by-step method. This technique has the advantage that one can derive directly the lowest-order terms in R^{-1}.

When we generate d in a step-by-step process, we are essentially producing d as a series of terms:

$$d(\mathbf{k};\mathbf{l}_1,\mathbf{l}_2) = \sum_{m=0}^{\infty} d^{(m)}(\mathbf{k};\mathbf{l}_1,\mathbf{l}_2) . \tag{2.17}$$

If we use (2.13) to transform (2.17) term by term, we will get

$$D(\mathbf{R};\mathbf{l}_1,\mathbf{l}_2) = \sum_{m=0}^{\infty} D^{(m)}(\mathbf{R};\mathbf{l}_1,\mathbf{l}_2) . \tag{2.18}$$

Basically we are going to use (2.16) and let n increase by successive integers starting from zero. Before each increase in n, we examine the remaining volume integral of $f-d$ for its nonanalytic parts. These are subtracted off, which just adds another term to the series (2.18) for $D(\mathbf{R};\mathbf{l}_1,\mathbf{l}_2)$. Then one can integrate by parts again, which just increases n by unity. This process is repeated, and hence the series for $D(\mathbf{R};\mathbf{l}_1,\mathbf{l}_2)$ is generated. Thus we rewrite (2.16) as

$$\begin{aligned} \frac{1}{2\pi^2} \int_{\mathrm{BZ}} d^3k\, e^{i\mathbf{k}\cdot\mathbf{R}} f = {} & \sum_{m=0}^{n-1} D^{(m)}(\mathbf{R};\mathbf{l}_1,\mathbf{l}_2) \\ & + \frac{1}{2\pi^2} \int_{\mathrm{BZ}} d^3k\, e^{i\mathbf{k}\cdot\mathbf{R}} \left(\frac{i\mathbf{R}\cdot\nabla_k}{R^2}\right)^n \Big[f - \sum_{m=0}^{n-1} d^{(m)}\Big] \\ & - \frac{1}{2\pi^2} \int_{\mathrm{OBZ}} d^3k\, e^{i\mathbf{k}\cdot\mathbf{R}} \left(\frac{i\mathbf{R}\cdot\nabla_k}{R^2}\right)^n \sum_{m=0}^{n-1} d^{(m)}(\mathbf{k};\mathbf{l}_1,\mathbf{l}_2) . \end{aligned} \tag{2.19}$$

In writing (2.19) we have assumed that $n \geq 1$. We already noted that the first subtraction must be

$$d^{(0)}(\mathbf{k};\mathbf{l}_1,\mathbf{l}_2)=Gk^{-2},$$
$$D^{(0)}(\mathbf{R};\mathbf{l}_1,\mathbf{l}_2)=GR^{-1}.$$

Some criteria must be agreed upon for deciding the terms to be subtracted. We have used the standard of subtracting out all terms in the volume integral for BZ in (2.19) which have a wave-vector power of k^{-2}. By this we mean wave-vector terms of dimension k^{-2}; the term k_x/k^3 thereby qualifies. For each value of n in (2.19), the term to be subtracted is denoted $C^{(n)}(\mathbf{k};\mathbf{l}_1,\mathbf{l}_2)$.

$C^{(n)}(\mathbf{k};\mathbf{l}_1,\mathbf{l}_2)=$ terms of order $(1/k^2)$ in

$$\left(\frac{i\mathbf{R}\cdot\nabla_k}{R^2}\right)^n \left[f-\sum_{m=0}^{n-1} d^{(m)}(\mathbf{k};\mathbf{l}_1,\mathbf{l}_2)\right]. \quad (2.20)$$

If no terms of order k^{-2} are present, then no subtraction is needed and $C^{(n)}=0$. The relationship between $C^{(m)}$ and $d^{(m)}$ is simply

$$C^{(m)}(\mathbf{k};\mathbf{l}_1,\mathbf{l}_2)=\left(\frac{i\mathbf{R}\cdot\nabla_k}{R^2}\right)^m d^{(m)}(\mathbf{k};\mathbf{l}_1,\mathbf{l}_2), \quad (2.21)$$

$$D^{(m)}(\mathbf{R};\mathbf{l}_1,\mathbf{l}_2)=\int_\infty \frac{d^3k}{2\pi^2} e^{i\mathbf{k}\cdot\mathbf{R}} C^{(m)}(\mathbf{k};\mathbf{l}_1,\mathbf{l}_2). \quad (2.22)$$

It should be emphasized that $C^{(m)}(\mathbf{k};\mathbf{l}_1,\mathbf{l}_2)$ is the important quantity. One never needs to determine $d^{(m)}(\mathbf{k};\mathbf{l}_1,\mathbf{l}_2)$ for $m>1$. The Fourier transforms $D^{(m)}(\mathbf{R};\mathbf{l}_1,\mathbf{l}_2)$ are found directly from $C^{(m)}$ in (2.22). This transform will always exist since we always chose $C^{(m)} \sim k^{-2}$. In contrast, note that the direct transform of $d^{(m)}(\mathbf{k};\mathbf{l}_1,\mathbf{l}_2)$ may not exist, but that this is unimportant for our calculation. Since $C^{(m)}$ is the important quantity, then in evaluating (2.19) one really would use the identity

$$\left(\frac{i\mathbf{R}\cdot\nabla_k}{R^2}\right)^n \sum_{m=0}^{n-1} d^{(m)}(\mathbf{k};\mathbf{l}_1,\mathbf{l}_2)=\left(\frac{i\mathbf{R}\cdot\nabla_k}{R^2}\right)^n d^{(0)}(\mathbf{k};\mathbf{l}_1,\mathbf{l}_2)$$
$$+\sum_{m=1}^{n-1}\left(\frac{i\mathbf{R}\cdot\nabla_k}{R^2}\right)^{n-m} C^{(m)}(\mathbf{k};\mathbf{l}_1,\mathbf{l}_2).$$

We hopefully can clarify how the $C^{(m)}$ are determined by listing the lowest two:

$$C^{(1)}=k^{-2} \text{ parts of } \left(\frac{i\mathbf{R}\cdot\nabla_k}{R^2}\right)[f-d^{(0)}(\mathbf{k};\mathbf{l}_1,\mathbf{l}_2)],$$

$$C^{(2)}=k^{-2} \text{ parts of } \left(\frac{i\mathbf{R}\cdot\nabla_k}{R^2}\right)^2[f-d^{(0)}]-\left(\frac{i\mathbf{R}\cdot\nabla_k}{R^2}\right)C^{(1)}.$$

We emphasize that by determining the $C^{(m)}(\mathbf{k};\mathbf{l}_1,\mathbf{l}_2)$ rather than $d^{(m)}(\mathbf{k};\mathbf{l}_1,\mathbf{l}_2)$, and by doing so in a step-by-step method, we do not alter the conclusion that these subtractions do not contribute any net surface integral.

In summary, we generate the $C^{(m)}$ by successively integrating by parts and subtracting out the non-analytic terms. These are Fourier transformed using (2.22) to obtain $D^{(m)}(\mathbf{R};\mathbf{l}_1,\mathbf{l}_2)$. These are added to give $D(\mathbf{R};\mathbf{l}_1,\mathbf{l}_2)$ in (2.18), and this is the evaluation of the integral in (2.1) since (2.16) is valid.

This method does produce the desired expansion in powers of R^{-1}. Since we always chose $C^{(m)}$ proportional to k^{-2}, then from (2.20) we see that dependence of $C^{(m)}$ is

$$C^{(m)}(\mathbf{k};\mathbf{l}_1,\mathbf{l}_2)\sim k^{-2}R^{-m}.$$

Therefore, the integral in (2.22) makes $D^{(m)}(\mathbf{R};\mathbf{l}_1,\mathbf{l}_2) \sim R^{-(m+1)}$ and the series (2.18) is a power series in R^{-1}. We can obtain the first few terms in the expansion for R^{-1} by evaluating first few of the $D^{(m)}(\mathbf{R};\mathbf{l}_1,\mathbf{l}_2)$.

We conclude this section by emphasizing that these results are quite general. They are valid for arbitrary position $\mathbf{l}_1$ and $\mathbf{l}_2$ of the two impurities in their unit cells.

III. EVALUATION OF THE POTENTIAL

A. Form of $f(k)$

In order to proceed with the actual calculations, it is necessary to have an explicit expression for the leading terms of $f(\mathbf{k})$ in powers of $\mathbf{k}$.[8] This can be obtained as follows: $W_\mu(\mathbf{l}_1,\mathbf{k})$ is given by

$$W_\mu(\mathbf{l}_1,\mathbf{k})=ik_\mu/k^2+A_\mu(\mathbf{l}_1)+ik_\lambda C_{\lambda\mu}(\mathbf{l}_1)+\cdots, \quad (3.1)$$

where A_μ and $C_{\lambda\mu}$ are expressible in terms of lattice sums, the exact forms of which are derived in Appendix B. The constants A_μ and $C_{\mu\lambda}$ are real.

The tensor $T_{\mu\nu}$ is of the form[4]

$$T_{\mu\nu}=k_\mu k_\nu/k^2-\tfrac{1}{3}\delta_{\mu\nu}+(a/\pi)^2 w_{\mu\nu\alpha\beta}k_\alpha k_\beta+\cdots, \quad (3.2)$$

which we denote as $T^{(0)}{}_{\mu\nu}+T^{(2)}{}_{\mu\nu}+\cdots$, the superscript denoting the order of the k dependence; the components of $w_{\mu\nu\alpha\beta}$ are given in I for the various cubic structures. Hence $G_{\mu\nu}$, which is the inverse of $\delta_{\mu\nu}+4\pi\alpha T_{\mu\nu}$, is given by

$$G_{\mu\nu}=G^{(0)}{}_{\mu\nu}-4\pi\alpha G^{(0)}{}_{\mu\lambda}T_{\lambda\kappa}G^{(0)}{}_{\kappa\nu}+O(k^4), \quad (3.3)$$

where $G^{(0)}{}_{\mu\nu}$ is the inverse of $\delta_{\mu\nu}+4\pi\alpha T^{(0)}{}_{\mu\nu}$. Specifically,

$$G^{(0)}{}_{\mu\nu}=[\delta_{\mu\nu}-4\pi\alpha k_\mu k_\nu/k^2(1+8\pi\alpha/3)](1-4\pi\alpha/3)^{-1}.$$

We note that $\mathbf{G}^{(0)}$ is diagonal in a coordinate system with $\mathbf{k}$ one of the axes; the $\hat{k}\hat{k}$ element of $G^{(0)}$ we shall call G. If we put together (3.1), (3.2), and (3.3), we get

$$f(k)=G/k^2+iGk_\mu[A_\mu(\mathbf{l}_2)-A_\mu(\mathbf{l}_1)]/k^2$$
$$+A_\mu G^{(0)}{}_{\mu\nu}A_\nu-4\pi\alpha G^2 k_\mu T^{(2)}{}_{\mu\nu}k_\nu/k^4$$
$$+G(k_\mu k_\nu/k^2)[C_{\mu\nu}(\mathbf{l}_1)+C_{\mu\nu}(\mathbf{l}_2)]+O(k). \quad (3.4)$$

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[8] Henceforth we simplify our notation by dropping the explicit reference to $\mathbf{l}_1$ and $\mathbf{l}_2$ in function arguments. Thus $f(\mathbf{k})$, $C^{(m)}(\mathbf{k})$, $d(\mathbf{k})$, and $D(\mathbf{R})$ are the previously defined functions, and still depend upon the positions $\mathbf{l}_1$ and $\mathbf{l}_2$ in the unit cells.

The first term is $O(k^{-2})$, the second $O(k^{-1})$, and the remaining three $O(k^0)$ by the conventions already established.

B. Integral Evaluation

We now return to (2.16). It is readily seen that $d^{(0)}(\mathbf{k})=G/k^2$, and hence

$$D^{(0)}(\mathbf{R})=G/R. \tag{3.5}$$

The leading term in $f-d^{(0)}(\mathbf{k})$ is $d^{(1)}(\mathbf{k})=iGk_\mu[A_\mu(\mathbf{l}_2)-A_\mu(\mathbf{l}_1)]/k^2$. Hence

$$C^{(1)}(\mathbf{k})=-\frac{1}{(kR)^2}G[A_\mu(\mathbf{l}_1)-A_\mu(\mathbf{l}_2)](\delta_{\mu\nu}-2k_\mu k_\nu/k^2)R_\nu. \tag{3.6}$$

Now

$$\frac{1}{2\pi^2}\int d^3k\, e^{i\mathbf{k}\cdot\mathbf{R}}\left(\delta_{\mu\nu}-\frac{2k_\mu k_\nu}{k^2}\right)\frac{1}{k^2}=\frac{R_\nu R_\mu}{R^3} \tag{3.7}$$

and therefore

$$D^{(1)}(\mathbf{R})=+GR_\mu[A_\mu(\mathbf{l}_1)-A_\mu(\mathbf{l}_2)]/R^3. \tag{3.8}$$

The next subtraction is $d^{(2)}(\mathbf{k})$, which consists of the remaining terms on the right-hand side of (3.4). Consider first $A_\mu(\mathbf{l}_1)G^{(0)}{}_{\mu\nu}A_\nu(\mathbf{l}_2)$; evaluate this in the coordinate system which diagonalizes $\mathbf{G}$. Using the representation (3.3), we find $A_\mu G^{(0)}{}_{\mu\nu}A_\nu=-(\epsilon-1)\times(\epsilon+2)A_\mu A_\nu k_\mu k_\nu/(k^2 3\epsilon)$ plus a term which is independent of k_ν. The constant term does not contribute to $D^{(2)}$, because one must differentiate to get $C^{(2)}$ from $d^{(2)}$. Carrying out the double gradient, and then integrating over k space, one obtains for the contribution of this term to $D^{(2)}(\mathbf{R})$

$$\frac{-(\epsilon-1)(\epsilon+2)}{3\epsilon}A_\mu(\mathbf{l}_1)\phi_{\mu\nu}(\mathbf{R})A_\nu(\mathbf{l}_2).$$

The last term involving $C_{\mu\nu}$ clearly contains exactly the same combination of k's, and hence yields

$$G[C_{\mu\nu}(\mathbf{l}_1)+C_{\mu\nu}(\mathbf{l}_2)]\phi_{\mu\nu}(\mathbf{R}).$$

The remaining contribution, involving $\mathbf{T}^{(2)}$, is a bit more complicated. We have shown the form of $\mathbf{T}^{(2)}$ in (3.2) for cubic crystals. Hence

$$k_\mu k_\nu T_{\mu\nu}{}^{(2)}/k^4=(2w_{44}-w_{11})-(2w_{44}-\tfrac{3}{2}w_{11})k_\mu{}^2k_\mu{}^2/k^4, \tag{3.9}$$

where $w_{11}=w_{xxxx}=w_{yyyy}=w_{zzzz}$, $w_{44}=w_{xyxy}=w_{xyyx}=\cdots$.[4] Again, the constant term plays no role. We carry out the double $\mathbf{k}$-space gradient, integrate over $\mathbf{k}$ space, and obtain for the contribution to $D^{(2)}(\mathbf{R})$

$$\tfrac{3}{2}R^{-3}(2w_{44}-\tfrac{3}{2}w_{11})4\pi\alpha G^2[5R_\mu{}^2R_\mu{}^2/R^4-3]. \tag{3.10}$$

The polynomial in square brackets in (3.10) will be recognized as a Kubic harmonic for the A_{1g} representation.[9] The potential must, of course, have this symmetry.

Now recall that

$$V(R)=\frac{q_1q_2}{R}\left[1-4\pi\alpha\sum_{m=0}^{n}D^{(m)}(R)+O(R^{-n-1})\right] \tag{3.11}$$

and that

$$G=\left(1+\frac{8\pi\alpha}{3}\right)^{-1}, \qquad 4\pi\alpha G=(\epsilon-1)/\epsilon. \tag{3.12}$$

Putting all these results together, we find

$$\begin{aligned}V(R)=q_1q_2\Bigg[&\frac{1}{\epsilon R}-\frac{(\epsilon-1)}{\epsilon}R_\mu\frac{A_\mu(\mathbf{l}_1)-A_\mu(\mathbf{l}_2)}{R^3}\\&+\frac{(\epsilon-1)^2}{\epsilon}A_\mu(\mathbf{l}_1)\phi_{\mu\nu}(\mathbf{R})A_\nu(\mathbf{l}_2)\\&-\frac{(\epsilon-1)^2}{\epsilon}\phi_{\mu\nu}(\mathbf{R})[C_{\mu\nu}(\mathbf{l}_1)+C_{\mu\nu}(\mathbf{l}_2)]\\&+\tfrac{3}{2}\left(\frac{\epsilon-1}{\epsilon}\right)^2\left(\frac{a}{\pi}\right)^2(\tfrac{3}{2}w_{11}-2w_{44})\\&\times(5R_\mu{}^2R_\mu{}^2/R^4-3)/R^3+O(R^{-4})\Bigg].\end{aligned} \tag{3.13}$$

C. Simplification

Although Eq. (3.13) looks fairly formidable, some simplification is possible if the impurities remain near the centers of their respective cells. Let the displacements from the centers be $\delta_\nu{}^1$ and $\delta_\nu{}^2$, respectively. Then (a) $A_\nu=\frac{1}{3}\delta_\nu+O(\delta^2)$ and (b) $C_{\mu\nu}=C\delta_{\mu\nu}+O(\delta^2)$. Thus, to order δ we have

$$V(R)=q_1q_2\left[\frac{1}{\epsilon R}-\left(\frac{\epsilon-1}{3\epsilon}\right)\frac{\mathbf{R}\cdot(\boldsymbol{\delta}^1-\boldsymbol{\delta}^2)}{R^3}+O(R^{-3})\right]. \tag{3.14}$$

The force at the center of the cell containing particle 2, say, is

$$\begin{aligned}F_\mu&\equiv q_1q_2\left[\frac{R_\mu}{\epsilon R^3}+\frac{\epsilon-1}{3\epsilon}\frac{R_\mu}{R^3}+O(R^{-3})\right]\\&=q_1q_2\tfrac{1}{3}(\epsilon+2)\frac{R_\mu}{\epsilon R^3}+O(R^{-3}).\end{aligned} \tag{3.15}$$

That is, we get immediately the well-known $\frac{1}{3}(\epsilon+2)$ local field correction: The local electric field at a site of cubic symmetry is $\frac{1}{3}(\epsilon+2)$ times the average internal field defined as $-\nabla(1/\epsilon R)$.

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[9] F. C. Von der Lage and H. A. Bethe, Phys. Rev. 71, 612 (1947).

IV. DISCUSSION

We have calculated some of the important contributions to the Coulomb interaction between two point charges in an atomic dielectric. This Coulomb potential has been shown to be a series—probably an asymptotic series—in powers of $1/R$. Furthermore, we have presented a method of evaluating the coefficients of the leading terms in this series. This method is easy to use and hard to justify. Thus Sec. II, in which we justify the method, is much longer than Sec. III, where we calculate the results. These results consist of the first few terms in the series in $1/R$ for the sc, bcc, and fcc lattices:

$$\begin{aligned} V(R)=q_1q_2\bigg[\frac{1}{\epsilon R}&-\frac{\epsilon-1}{\epsilon}[A_\mu(\mathbf{l}_1)-A_\mu(\mathbf{l}_2)]\frac{R_\mu}{R^2}\\ &+\frac{(\epsilon-1)^2}{\epsilon}A_\mu(\mathbf{l}_1)\phi_{\mu\nu}(\mathbf{R})A_\nu(\mathbf{l}_2)\\ &-\frac{\epsilon-1}{\epsilon}[C_{\mu\nu}(\mathbf{l}_1)+C_{\mu\nu}(\mathbf{l}_2)]\phi_{\mu\nu}(\mathbf{R})\\ &+\tfrac{3}{2}(\tfrac{3}{2}w_{11}-2w_{44})\left(\frac{\epsilon-1}{\epsilon}\right)^2\frac{1}{R^3}\left[5\left(\frac{R_\mu{}^2R_\mu{}^2}{R^2}\right)-3\right]\\ &\times(a/\pi)^2+O(1/R^4)\bigg]. \quad (4.1)\end{aligned}$$

This result is interesting because some of the terms are intuitively reasonable, while others are apparently not derivable by insight alone. For example, the $\mathbf{A}\cdot\mathbf{R}/R^3$ terms are just a charge-dipole interaction. If the impurities $\mathbf{l}_1$ are located at a position with inversion symmetry, the coefficient $\mathbf{A}(\mathbf{l}_1)$ vanishes. However, if the impurity is located at an off-center position within the unit cell, then the impurity is nearer to some host atoms than to others. The nearby ones get polarized more. This gives the unit cell containing the impurity—indeed, the entire crystal region surrounding the impurity—a net polarization moment. This provides the dipole moment for the charge-dipole interaction with the other impurity charge. The coefficient $\bar{A}_\mu=(\epsilon-1)A_\mu$ is just the value of this dipole moment. We can also see that the term

$$\bar{A}_\mu\phi_{\mu\nu}(\mathbf{l})\bar{A}_\nu/\epsilon$$

is just the dipole-dipole interaction between the two polarization moments caused by the two charges. The approximation $A_\mu=\frac{1}{3}\delta_\mu$ only applies when the impurity is displaced a small distance $\boldsymbol{\delta}$ from a position of cubic symmetry.

Similarly, most positions of the impurity within the unit cell will cause the polarization field around the impurity to have a quadrupole moment. This is the origin of the term

$$[(\epsilon-1)/\epsilon]C_{\mu\nu}\phi_{\mu\nu}(\mathbf{R}),$$

which is just the interaction of this quadrupole with the other impurity charge. The value of the quadrupole moment is $\bar{C}_{\mu\nu}=(\epsilon-1)C_{\mu\nu}$. However, if the impurity is at a position of cubic symmetry, there is no quadrupole moment, as was noted above; in this case $C_{\mu\nu}=C\delta_{\mu\nu}$ and the entire term vanishes.

The polarization cloud surrounding the impurity may also have higher moments, and these induce interactions with higher powers of R. Note that for an impurity at $\mathbf{l}_1$, all of these induced polarization moments are derivable from the wave-vector expansion of $W_\mu(\mathbf{l}_1,\mathbf{k})$. It appears in the definition of

$$f(\mathbf{k})=W_\mu(\mathbf{l}_1,\mathbf{k})G_{\mu\nu}(\mathbf{k})W_\nu(\mathbf{l}_2,-\mathbf{k})$$

that the W's describe the formation of the two polarization clouds around each of these two impurities, while $G_{\mu\nu}$ describes how these two polarization clouds interact.

The terms involving the dipole A_μ and the quadrupole $C_{\mu\nu}$ could perhaps have been derived by intuition without having to go through our complicated analysis. On the other hand, we do not see how the term involving $w_{\alpha\beta}$, and its proportionality to a Kubic harmonic, could have been derived except by starting as we did from Eq. (2.1). These terms arise from spatial dispersion, i.e., from the wave-vector dependence of the dielectric response function. This fact is easily demonstrated by showing how $G_{\mu\nu}(\mathbf{k})$, and its wave-vector dependence, enter into the definition of the dielectric response function $\epsilon(\mathbf{q},\omega)$. Let us consider how an applied electric field

$$\mathbf{E}(\mathbf{l},t)=\mathbf{E}_0e^{i(\mathbf{k}\cdot\mathbf{l}-\omega t)}$$

propagates through the lattice. At each lattice site $\mathbf{l}$ a polarization

$$\mathbf{P}(\mathbf{l},t)=\mathbf{P}_0e^{i(\mathbf{k}\cdot\mathbf{l}-\omega t)}$$

will be induced. By considering the local electric field at each lattice site, one can show that the polarization amplitude is obtained by solving the equation[10]

$$\mathbf{P}_0=\boldsymbol{\alpha}\cdot[\mathbf{E}_0-4\pi\mathbf{T}(\mathbf{k})\cdot\mathbf{P}_0].$$

From (2.5) we see that this has the solution

$$\mathbf{P}_0=\mathbf{G}(\mathbf{k})\cdot\boldsymbol{\alpha}\cdot\mathbf{E}_0.$$

This result, along with Maxwell's equation,

$$\mathbf{E}_0+4\pi\mathbf{P}_0=-(c/\omega)^2\mathbf{k}\times(\mathbf{k}\times\mathbf{E}_0),$$

defines the normal modes of the point-dipole solid, which in turn give the dielectric response function. Only by starting from (2.1) would one know how to properly include the spatial dispersion characteristics of the solid.

Equations similar to (2.1) were given in I for the charge-dipole and dipole-dipole interactions between

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[10] G. D. Mahan, J. Chem. Phys. 43, 1569 (1965).

impurities. Although we have not analyzed the asymptotic expansions of these interactions in any detail, it is easy to see that they could be evaluated by the same method. For example, by examining the function which is equivalent to $f(\mathbf{k})$ for these two types of interactions, one sees immediately that the surface integrals vanish as they did for $f(\mathbf{k})$ in Eq. (2.11). Thus it appears that the asymptotic series for the charge-dipole and dipole-dipole interactions between impurities could also be found by just subtracting and Fourier-transforming the nonanalytic parts of the interaction.

Our calculation assumed that the host dielectric was composed of point ions, which when polarized are point dipoles. Within the framework of this point-dipole model, we have been able to calculate exactly the leading terms in the asymptotic expansion of the Coulomb interaction between impurity charges. Although this model is a highly idealized simplification of a real dielectric, it is employed widely to calculate the properties of real materials. This is because point-dipole calculations are relatively easy to perform, while computations are very much harder for actual solids with finite and overlapping charge distributions. We have adopted the attitude that, as long as this model is going to be employed so extensively, it is worthwhile to deduce its properties as accurately as possible. We feel that our calculation of the asymptotic properties of the Coulomb potential is a contribution towards understanding this model.

It is interesting to speculate how our results would be altered in a real dielectric with finite charge distributions. It is reasonable to conclude that the main features of our results would pertain to the potential in an actual dielectric. An impurity charge off center in the atomic cell would still cause the induced polarization field to have a dipole, quadrupole, and higher moments. One would still get the same type of interaction terms between these induced moments, and only the magnitude of the moments are altered by the properties of the real dielectric. Similarly, in a real insulator the wave-vector dependence of the dielectric response function will be different than in the point-dipole model. The term proportional to $w_{\alpha\beta}$ which arose from the wave-vector dependence of $G_{\mu\nu}(\mathbf{k})$ will be different in the real dielectric. On the other hand, since the general form of $T_{\mu\nu}(\mathbf{k})$ is fixed by group-theoretical arguments, then only the values of the parameters w_{11}, w_{12}, w_{44} can change. The basic form of the interaction term in (4.1) should be the same, and only the coefficient $w_{\alpha\beta}$ should change. This discussion suggests that in a real insulator, the asymptotic expansion for the Coulomb potential should have the same form as we have derived for the point-dipole model: The only change is that the parameters A_μ, $C_{\mu\nu}$, and $w_{\alpha\beta}$ will have different values. However, Sham's calculation[6] indicates that additional terms may occur which arise from the effect of correlation and exchange.

APPENDIX A: TWO ATOMS PER CELL

Equations (2.1) and (2.2) only apply to solids with one atom per unit cell. In this Appendix we derive the equivalent relations for solids with two atoms per unit cell. This is a fairly important case, since it includes lattice structures such as alkali halides, diamond, and zinc blende.

We arbitrarily select one sublattice and designate its quantities by a superscript (1), e.g., its atoms have a polarizability $\alpha^{(1)}$. The atomic positions of the sublattice are at $\mathbf{l}$. The equivalent to (2.3), which is the wave-vector transform of the charge-dipole interaction between an impurity at $\mathbf{l}_0$ and this sublattice, is

$$W^{(1)}{}_{\nu}(\mathbf{l}_0,\mathbf{k})=\frac{V_0}{4\pi}\sum_{l} e^{i\mathbf{k}\cdot(\mathbf{l}-\mathbf{l}_0)}\frac{(\mathbf{l}-\mathbf{l}_0)_\nu}{(\mathbf{l}-\mathbf{l}_0)^3}. \tag{A1}$$

Similarly, the functions associated with the second sublattice are designated by a superscript (2). Here the atoms positions are at $(\mathbf{l}+\boldsymbol{\tau})$, where $\boldsymbol{\tau}$ is a nonprimitive translation. Thus one has to define an additional transform

$$W^{(2)}{}_{\nu}(\mathbf{l}_0,\mathbf{k})=\frac{V_0}{4\pi}\sum_{l} e^{i\mathbf{k}\cdot(\mathbf{l}+\tau-\mathbf{l}_0)}\frac{(\mathbf{l}+\boldsymbol{\tau}-\mathbf{l}_0)_\gamma}{(\mathbf{l}+\boldsymbol{\tau}-\mathbf{l}_0)^3}, \tag{A2}$$

where V_0 is the volume of a unit cell, and $\alpha^{(1)}$ and $\alpha^{(2)}$ are the atomic polarizabilities per unit cell volume.

One must also enlarge the number of wave-vector transforms of the dipole-dipole interaction. Let us define

$$T^{(i,j)}{}_{\mu\nu}(\mathbf{k})=\frac{V_0}{4\pi}\sum_{\mathbf{l}^{(j)}} \exp[i\mathbf{k}\cdot(\mathbf{l}^{(i)}-\mathbf{l}^{(j)})]\phi_{\mu\nu}(\mathbf{l}^{(i)}-\mathbf{l}^{(j)}), \tag{A3}$$

where $\mathbf{l}^{(1)}=\mathbf{l}$, $\mathbf{l}^{(2)}=\mathbf{l}+\boldsymbol{\tau}$. When we write out the four possibilities (i, $j=1$, 2), there are only two different transforms. In accord with our prior usage,[11] we define the equivalent $\mathbf{T}^{(e)}(\mathbf{k})$ and unequivalent $\mathbf{T}^{(i)}(\mathbf{k})$ sums

$$\begin{aligned} T^{(e)}{}_{\mu\nu}(\mathbf{k})&=T^{(1,1)}{}_{\mu\nu}(\mathbf{k})=T^{(2,2)}{}_{\mu\nu}(\mathbf{k})=\frac{V_0}{4\pi}\sum_{l\neq0} e^{i\mathbf{k}\cdot\mathbf{l}}\phi_{\mu\nu}(\mathbf{l}),\\ T^{(i)}{}_{\mu\nu}(\mathbf{k})&=T^{(2,1)}{}_{\mu\nu}(\mathbf{k})=T^{(1,2)}{}_{\mu\nu}(-\mathbf{k})\\ &=\frac{V_0}{4\pi}\sum_{l} e^{i\mathbf{k}\cdot(\mathbf{l}-\tau)}\phi_{\mu\nu}(\mathbf{l}-\boldsymbol{\tau}). \end{aligned} \tag{A4}$$

Since for all crystal structures the *sub*lattice has inversion symmetry, then $T^{(e)}{}_{\mu\nu}(\mathbf{k})=T^{(e)}{}_{\mu\nu}(-\mathbf{k})$. But only for structures in which $2\boldsymbol{\tau}=\mathbf{l}$ does $T^{(i)}{}_{\mu\nu}(\mathbf{k})=T^{(i)}{}_{\mu\nu}(-\mathbf{k})$. Thus there are lattices where $T^{(e)}{}_{\mu\nu}(\mathbf{k})$ is invariant under $\mathbf{k}$ reversal, although $T^{(i)}{}_{\mu\nu}(\mathbf{k})$ is not; for example, diamond.

The functions $G_{\mu\nu}(\mathbf{k})$ define how polarization is propagated through the lattice. For the case of two

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[11] G. D. Mahan, J. Chem. Phys. 41, 2930 (1964).

atoms per cell, we must evaluate four functions $G^{(i,j)}{}_{\mu\nu}(\mathbf{k})$, where i, $j=1$, 2. These are defined as the probability that polarization starts on the (i) type of atom and travels to the (j) type. Hence these functions have the equations

$$\begin{aligned}\mathbf{G}^{(1,1)}(\mathbf{k})&=\mathbf{I}-4\pi\mathbf{T}^{(e)}(\mathbf{k})\cdot\boldsymbol{\alpha}^{(1)}\cdot\mathbf{G}^{(1,1)}(\mathbf{k})\\&\qquad-4\pi\mathbf{T}^{(i)}(\mathbf{k})\cdot\boldsymbol{\alpha}^{(2)}\cdot\mathbf{G}^{(2,1)}(\mathbf{k}),\\\mathbf{G}^{(2,1)}(\mathbf{k})&=-4\pi\mathbf{T}^{(e)}(\mathbf{k})\cdot\boldsymbol{\alpha}^{(2)}\cdot\mathbf{G}^{(2,1)}(\mathbf{k})\\&\qquad-4\pi\mathbf{T}^{(i)}(\mathbf{k})\cdot\boldsymbol{\alpha}^{(1)}\cdot\mathbf{G}^{(1,1)}(\mathbf{k}),\\\mathbf{G}^{(1,2)}(\mathbf{k})&=-4\pi\mathbf{T}^{(e)}(\mathbf{k})\cdot\boldsymbol{\alpha}^{(1)}\cdot\mathbf{G}^{(1,2)}(\mathbf{k})\\&\qquad-4\pi\mathbf{T}^{(i)}(\mathbf{k})\cdot\boldsymbol{\alpha}^{(2)}\cdot\mathbf{G}^{(2,2)}(\mathbf{k}),\\\mathbf{G}^{(2,2)}(\mathbf{k})&=\mathbf{I}-4\pi\mathbf{T}^{(e)}(\mathbf{k})\cdot\boldsymbol{\alpha}^{(2)}\cdot\mathbf{G}^{(2,2)}(\mathbf{k})\\&\qquad-4\pi\mathbf{T}^{(i)}(\mathbf{k})\cdot\boldsymbol{\alpha}^{(i)}\cdot\mathbf{G}^{(1,2)}(\mathbf{k}).\end{aligned}\tag{A5}$$

These equations can be solved to obtain the four tensors $\mathbf{G}^{(i,j)}(\mathbf{k})$. After this has been done, one can evaluate the Coulomb interaction between two impurity charges q_a, q_b at $\mathbf{l}_a$ and $\mathbf{l}_b$, with $\mathbf{R}=\mathbf{l}_a-\mathbf{l}_b$. This result is

$$V(\mathbf{R})=q_aq_b\left[\frac{1}{R}-\frac{1}{2\pi^2}\int_{\mathrm{BZ}}d^3k\,e^{i\mathbf{k}\cdot\mathbf{R}}\bar{f}(\mathbf{k})\right],\tag{A6}$$

where

$$\begin{aligned}\bar{f}(\mathbf{k})=4\pi\sum_{i=1;\,j=1}^{2}&\mathbf{W}^{(i)}(\mathbf{l}_a,\mathbf{k})\cdot\boldsymbol{\alpha}^{(i)}\\&\cdot\mathbf{G}^{(i,j)}(\mathbf{k})\cdot\mathbf{W}^{(j)}(\mathbf{l}_b,-\mathbf{k}).\end{aligned}\tag{A7}$$

We have denoted the above function by $\bar{f}$, since it differs in normalization from the f in (2.1) by a factor of $4\pi\alpha$.

These equations can be simplified if one just wishes to evaluate the Coulomb interaction in (A6). One does not need to evaluate the four tensor functions $\mathbf{G}^{(i,j)}(\mathbf{k})$. Instead, let us define two vector functions $U_\nu{}^{(i)}$ for $i=1$, 2:

$$U^{(i)}{}_\mu(\mathbf{l}_b,\mathbf{k})=\sum_{j=1}^{2}G_{\mu\nu}{}^{(i,j)}(\mathbf{k})W^{(j)}{}_\nu(\mathbf{l}_b,-\mathbf{k}).\tag{A8}$$

If we take the four equations (A5) and insert them into (A8), we generate the equations

$$\begin{aligned}U^{(1)}{}_\mu(\mathbf{l}_b,\mathbf{k})&=W^{(1)}{}_\mu(\mathbf{l}_b,-\mathbf{k})-4\pi T^{(e)}{}_{\mu\nu}(\mathbf{k})\alpha^{(1)}{}_{\nu\lambda}U^{(1)}{}_\lambda\\&\qquad-4\pi T^{(i)}{}_{\mu\nu}(\mathbf{k})\alpha^{(2)}{}_{\nu\lambda}U^{(2)}{}_\lambda,\\U^{(2)}{}_\mu(\mathbf{l}_b,\mathbf{k})&=W^{(2)}{}_\mu(\mathbf{l}_b,-\mathbf{k})\\&\qquad-4\pi T^{(e)}{}_{\mu\nu}(\mathbf{k})\alpha^{(2)}{}_{\nu\lambda}U^{(2)}{}_\lambda(\mathbf{l}_b,k)\\&\qquad-4\pi T^{(i)}{}_{\mu\nu}(\mathbf{k})\alpha^{(1)}{}_{\nu\lambda}U^{(1)}{}_\lambda.\end{aligned}\tag{A9}$$

Furthermore, from (A7) and (A8) we see that

$$\bar{f}=4\pi\sum_{j=1}^{2}\mathbf{W}^{(j)}(\mathbf{l}_a,\mathbf{k})\cdot\boldsymbol{\alpha}^{(j)}\cdot\mathbf{U}^{(j)}(\mathbf{l}_b,\mathbf{k}).\tag{A10}$$

One only needs to solve for the two vector functions in (A9) in order to obtain (A10). This should be easier than solving for the four tensor functions $\mathbf{G}^{(i,j)}$.

A further simplification is possible in the case that all of the atoms have the same polarizability, $\alpha^{(1)}=\alpha^{(2)}$. Then it is convenient to introduce

$$\begin{aligned}U^{(\pm)}{}_\mu(l_b,k)&=U^{(1)}{}_\mu\pm U^{(2)}{}_\mu,\\W^{(\pm)}{}_\mu(l_a,k)&=W^{(1)}{}_\mu\pm W^{(2)}{}_\mu,\\T^{(\pm)}{}_{\mu\nu}&=T^{(e)}{}_{\mu\nu}\pm T^{(i)}{}_{\mu\nu}.\end{aligned}$$

By adding and subtracting (A9), we get that

$$\begin{aligned}U^{(\pm)}{}_\mu&=W^{(\pm)}{}_\mu-4\pi T^{(\pm)}{}_{\mu\nu}\alpha_{\nu\lambda}U^{(\pm)}{}_\lambda,\\\bar{f}&=2\pi[\mathbf{W}^{(+)}\boldsymbol{\alpha}\mathbf{U}^{(+)}+\mathbf{W}^{(-)}\boldsymbol{\alpha}\mathbf{U}^{(-)}].\end{aligned}\tag{A11}$$

In (A11), one just needs to solve a single vector equation for $\mathbf{U}^{(+)}$ and one for $\mathbf{U}^{(-)}$; the pair of equations (A9) become uncoupled when $\boldsymbol{\alpha}^{(1)}=\boldsymbol{\alpha}^{(2)}$.

We have not carried out any detailed calculations for the two-atom-per-cell case.

APPENDIX B: LATTICE SUMS

The technique we use for dealing with lattice sums is due to Born and Bradburn[12] and some of the formulas are given also by Cohen and Keffer.[13]

$W_\mu(\mathbf{l}_1,\mathbf{k})$ is given by Eq. (2.3), and can be written

$$\frac{4\pi}{V_0}W_\mu(\mathbf{l}_1,\mathbf{k})=\frac{1}{i}\frac{\partial}{\partial k_\mu}\sum_{l}e^{i\mathbf{k}\cdot(\mathbf{l}-\mathbf{l}_1)}|\mathbf{l}-\mathbf{l}_1|^{-3}.\tag{B1}$$

The sum in (B1) is like the S_3 of Cohen and Keffer, except that $\mathbf{l}_1$ is not a lattice vector. Nevertheless, the Born-Bradburn techniques are still applicable; one must merely use the generalized Ewald transformation formula given by Born and Huang,[14] instead of the simple one used by Born and Bradburn. We find that

$$\begin{aligned}\frac{4\pi}{V_0}W_\mu(\mathbf{l}_1,\mathbf{k})=i\frac{\partial}{\partial k_\mu}\Bigg\{&\frac{\eta^3}{\Gamma(\frac{3}{2})}\sum_{\mathbf{l}}e^{i\mathbf{k}\cdot(\mathbf{l}-\mathbf{l}_1)}\phi_{1/2}(\eta|\mathbf{l}-\mathbf{l}_1|^2)\\&+\frac{\pi^{3/2}}{V_0\Gamma(\frac{3}{2})\eta^3}\sum_{\mathbf{b}}e^{-i\mathbf{b}\cdot\mathbf{l}_1}\phi_{-1}(|\mathbf{b}-\mathbf{k}|^2/4\eta)\Bigg\}.\end{aligned}\tag{B2}$$

Here η is the parameter which regulates the relative contribution of the direct and reciprocal lattice sums. The total sum is independent of η. The $\mathbf{b}$ vectors are 2π times reciprocal lattice vectors, V_0 is the volume of a unit cell, and the ϕ functions are as given by Refs. 12 and 13. Thus

$$\begin{aligned}\frac{4\pi}{V_0}W_\mu(\mathbf{l}_1,\mathbf{k})=&\frac{\eta^3}{\Gamma(\frac{3}{2})}\sum_{\mathbf{l}}e^{i\mathbf{k}\cdot(\mathbf{l}-\mathbf{l}_1)}(\mathbf{l}-\mathbf{l}_1)_\mu\phi_{1/2}(\eta^2|\mathbf{l}-\mathbf{l}_1|^2)\\&+\frac{1}{i}\frac{\pi^{3/2}}{V_0\Gamma(\frac{3}{2})}\sum_{\mathbf{b}}e^{-i\mathbf{b}\cdot\mathbf{l}_1}\frac{(\mathbf{b}-\mathbf{k})_\mu}{2\eta^2}\phi_0(|\mathbf{b}-\mathbf{k}|^2/4\eta^2).\end{aligned}\tag{B3}$$

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[12] M. Born and M. Bradburn, Proc. Camb. Phil. Soc. **39**, 113 (1943).

[13] M. H. Cohen and F. Keffer, Phys. Rev. **19**, 1128 (1955).

[14] M. Born and K. Huang, *Dynamical Theory of Crystal Lattice* (Clarendon Press, Oxford, England, 1954), p. 251.

If we now expand in powers of $\mathbf{k}$, we get, from the $\mathbf{b}=0$ term of (B3),

$$\frac{2\pi^{3/2}}{\Gamma(\frac{3}{2})V_0}\frac{i\mathbf{k}}{k^2}[1-k^2/4\eta^2+O(k^4)]. \tag{B4}$$

From the direct-lattice sum we get

$$\frac{\eta^3}{\Gamma(\frac{3}{2})}\Big(\sum_{\mathbf{l}}(\mathbf{l}-\mathbf{l}_1)_\mu\phi_{1/2}(\eta^2|\mathbf{l}-\mathbf{l}_1|^2) +ik_\nu\sum_{l}(\mathbf{l}-\mathbf{l}_1)_\nu(\mathbf{l}-\mathbf{l}_1)_\mu\phi_{1/2}(\eta^2|\mathbf{l}-\mathbf{l}_1|^2)+\cdots\Big), \tag{B5}$$

while the reciprocal-lattice sum gives

$$\frac{2\pi^{3/2}}{V_0\Gamma(\frac{3}{2})}\sum_{\mathbf{b}\neq 0}e^{-i\mathbf{b}\cdot\mathbf{l}_1}\frac{(\mathbf{b}-\mathbf{k})_\mu}{|\mathbf{b}-\mathbf{k}|^2}e^{-|\mathbf{b}-\mathbf{k}|^2/4\eta^2}. \tag{B6}$$

If we further expand (B6) in powers of k_μ, we obtain

$$\frac{2\pi^{3/2}}{iV_0\Gamma(\frac{3}{2})}\sum_{\mathbf{b}}e^{-i\mathbf{b}\cdot\mathbf{l}_1-b^2/4\eta^2} \times\left[\frac{b_\mu}{b^2}-\frac{k_\mu}{b^2}+\frac{2b_\mu b_\alpha k_\alpha}{b^4}+\frac{b_\mu b_\alpha k_\alpha}{2b^2\eta^2}+O(k^2)\right]. \tag{B7}$$

Collecting powers of k_μ, we finally get Eq. (3.1), where

$$A_\mu(\mathbf{l}_1)=\frac{V_0}{2\pi^{3/2}}\eta^3\sum_{\mathbf{l}}(\mathbf{l}-\mathbf{l}_1)_\mu\phi_{1/2}(\eta^2|\mathbf{l}-\mathbf{l}_1|^2) -i\sum_{\mathbf{b}\neq 0}e^{-i\mathbf{b}\cdot\mathbf{l}_1}e^{-b^2/4\eta^2}b_\mu/b^2, \tag{B8}$$

$$C_{\mu\nu}(\mathbf{l}_1)=\frac{\eta^3V_0}{2\pi^{3/2}}\sum_{\mathbf{l}}(\mathbf{l}-\mathbf{l}_1)_\mu(\mathbf{l}-\mathbf{l}_1)_\nu\phi_{1/2}(\eta|\mathbf{l}-\mathbf{l}_1|^2) +\sum_{\mathbf{b}\neq 0}e^{-i\mathbf{b}\cdot\mathbf{l}_1}e^{-b^2/4\eta^2}[\delta_{\mu\nu}-b_\mu b_\nu(2/b^2-1/2\eta^2)]/b^2. \tag{B9}$$

It is clear by inspection that $\mathbf{A}(\mathbf{l}_1)$ vanishes if $\mathbf{l}_1$ lies as the exact center of a cell. $C_{\mu\nu}(\mathbf{l}_1)$ does not vanish in this circumstance, but is a multiple of the unit tensor.

If we write the deviation of $\mathbf{l}_1$ from the center of a cell as $\boldsymbol{\delta}$, then for $\boldsymbol{\delta}$ small, the deviation of $C_{\mu\nu}$ from its central value is $O(\delta^2)$. If we expand A_μ in powers of $\boldsymbol{\delta}$, however, we find

$$A_\mu=A\delta_\mu, \tag{B10}$$

where

$$A=\frac{V_0\eta^3}{2\pi^{3/2}}\Big(-\sum_{\mathbf{l}}\phi_{1/2}(\eta^2|\mathbf{l}-\mathbf{l}_0|^2) +\tfrac{2}{3}\eta^2\sum_{\mathbf{l}}(l-l_0)^2\phi_{3/2}(\eta^2|l-l_0|^2)\Big) -\tfrac{1}{3}\sum_{\mathbf{b}\neq 0}e^{-i\mathbf{b}\cdot\mathbf{l}_0}e^{-b^2/4\eta^2}. \tag{B11}$$

Here, $\mathbf{l}_0$ is a vector to a point at the center of a cell. This rather complicated expression is actually equal to $\frac{1}{3}$. This can be shown by applying the general Ewald transformation exactly as Cohen and Keffer applied the simple Ewald transformation to dipole wave sums.

The other lattice sums which enter the present problem are those defining $T_{\mu\nu}$. They are not discussed further here, since they do not involve any interstitial atoms, and have already been discussed in the literature.

APPENDIX C: EVALUATION OF SOME INTEGRALS

Integrals of the type

$$F_n(\mathbf{R})=\int e^{i\mathbf{k}\cdot\mathbf{R}}k_\mu{}^n/k^{n+2}d^3k \tag{C1}$$

appear in the foregoing; the subscripts refer to crystal axes which may have nothing to do with the direction of $\mathbf{R}$. Such integrals can be evaluated as follows. In cylindrical coordinates, with the k_μ axis as z axis, and $K^2=k^2-k_\mu{}^2$, we have

$$F_n(\mathbf{R})=2\int_0^\infty\cos(k_\mu R_\mu)k_\mu{}^n dk_\mu \times\int_0^\infty\frac{KdK}{(k_\mu{}^2+K^2)^{n/2+1}}\int_0^{2\pi}ie^{iK\rho\cos\phi}d\phi, \tag{C2}$$

where $\rho^2=R^2-R_\mu{}^2$, and here, repeated indices do not mean a sum. The ϕ integral is a Bessel function, so that

$$F_n(\mathbf{R})=4\pi\int_0^\infty\cos(k_\mu R_\mu)k_\mu{}^n dk_\mu\int_0^\infty\frac{J_0(K\rho)}{(k_\mu{}^2+K^2)^{n/2+1}}KdK. \tag{C3}$$

Now the K integral yields[15]

$$(\rho/2k_\mu)^{n/2}K_{-n/2}(k_\mu\rho)/\Gamma(\tfrac{1}{2}n+1),$$

where $K_{-n/2}$ is the Bessel function of imaginary argument. Hence[16]

$$F_n(\mathbf{R})=\frac{4\pi}{\Gamma(\frac{1}{2}n+1)}(\tfrac{1}{2}\rho)^{n/2} \times\int_0^\infty\cos(k_\mu R_\mu)K_{-n/2}(k_\mu\rho)k_\mu{}^{n/2}dk_\mu =\frac{2\pi^{3/2}}{R}\frac{\Gamma(\frac{1}{2}n+\frac{1}{2})}{\Gamma(\frac{1}{2}n+1)}\left(1-\frac{R_\mu{}^2}{R^2}\right)^{n/2}, \tag{C4}$$

which is the result we need.

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[15] *Higher Transcendental Functions*, edited by A. Erdelyi (McGraw-Hill Book Co., New York, 1953), Vol. 2, p. 96.

[16] *Table of Integral Transforms*, edited by A. Erdelyi (McGraw-Hill Book Co., New York, 1954), Vol. 1, p. 49.